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AN EMPIRICAL FORMULATION OF THERMIONIC CONVERTER PERFORMANCE AS A FUNCTION OF ELECTRODE EMISSION PROPERTIES

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SUMMARY

A simple empirical equation giving the emission current density in terms of the cesium atom arrival rate and the electrode surface temperature is presented in this report. The Swanson, Strayer correlation for the maximum change in work function due to cesium adsorption is compared with field-emission data for some refractory metals. Oxygen effects are discussed.

INTRODUCTION

Thermionic converter analysis would be facilitated by a simple equation giving the electron emission current density J explicitly in terms of the electrode temperature T and the cesium particle arrival rate μ.

The results should, of course, agree with existing experimental data. Also the expression must predict J where no experimental data exist (e.g., for μ larger than in existing data and for untried electrode materials).

Even with the progress that has been made toward developing a general understanding of the effect of alkali metal adsorption on the electron work function, there is as yet no basic calculation of J as a function of T and μ. A number of semiempirical correlations (refs. 1 to 8), however, relate J to T and cesium gas properties. Usually the most tested and discussed of these are the classical atomic physics approach of Rasor and Warner (ref. 3) and the molecular chemistry formulation of Levine and Gyftopoulos (ref. 5).

Although both works have in some instances agreed with experiments, some basic assumptions of the Rasor, Warner and the Levine, Gyftopoulos correlations have been questioned (refs. 9 to 12), both disagree with some experiments (refs. 13 to 18). While these works at least can be used as complex data fits, straightforward empiricism can yield a simpler formulation.

Presented in this report is a simple, accurate converter physics formulation. The result is an explicit equation for J in terms of the bare work function φ_0, μ, and T. Since the derivation is purely empirical, agreement with experiment is improved. This formulation is used to analyze the performance of an oxygenated cesiated diode.

THE CATHODE

A good deal of information has been obtained empirically about converter cathode surfaces. For example, Breitwieser's experiments (ref. 19) yield nearly linear plots of ln J against ln P_{Cs}, cesium pressure, for constant T's. In addition, the plots of effective work function φ against T/T_r are nearly independent of T_r for experiments with $\mu = P_{Cs}/\sqrt{2\pi mkT}$, where P_{Cs} is the vapor pressure of cesium at T_r. And φ, T/T_r relations are linear for $2.3 \leq \varphi \leq 3.3$, the useful range for power producing thermionic converters. Further, experimenters (ref. 20) provided methods to reduce data scatter below that of 0.1 eV for φ, T/T_r plots.

Houston (ref. 17) using the "plasma anode" technique, has shown that each of the metals tungsten (W), palladium (Pd), ruthenium (Ru), osmium (Os), rhodium (Rh), iridium (Ir), and platinum (Pt) gives its own curve when φ is plotted against T/T_r for cesium (Cs) reservoir temperatures of 414, 434, and 454 K. These data are nearly linear for $2.3 \leq \varphi \leq 3.3$ eV and can be correlated by

$$\varphi = \frac{aT}{T_r} + b \tag{1}$$

$$J = 120\, T^2 \exp\left[-(aT/T_r + b)kT\right] \tag{2}$$

where a and b are to be determined empirically. Equations (1) and (2) apply where $\mu = P_{Cs}/\sqrt{2\pi mkT_r}$. Generalizations will be discussed in the section THE CATHODE.

If experimental data were available for all possible cathode surfaces, it would be merely necessary to fit for a and b. But, of course, such data are not available; so it is desirable to write empirically a and also b in terms of cathode properties. The bare work function φ_0 is a property which distinguishes cathode surfaces; so it is desirable to find $a(\varphi_0)$ and $b(\varphi_0)$.

Since the work function of a surface is very sensitive to surface condition such as contamination, history of heat treatment, sputtering, and bulk impurity content, the following criteria should be satisfied by experiments designed to determine $a(\varphi_0)$ and $b(\varphi_0)$ empirically. First, it is desirable to have all samples measured in the same tube. Second, the bare work function should be obtained in the same tube that the cesiated data

are obtained. Houston (ref. 17) and Wilson (refs. 21 and 22) obtained experimental data in this manner. Their results are not in agreement, however. Houston offers at least plausible arguments for some disagreement in Wilson's results; so Houston's data are used in this report. The values of a and b determined empirically can, of course, be improved as progress is made experimentally.

A least-mean-square straight-line fit was used to determine a and b for the metals reported by Houston (except for which there were insufficient data). The results are given in table I. The rms deviations of $aT/T_r + b$ from the data Δ_{rms} and the magnitudes of the maximum deviations Δ_{max} are seen to be quite small.

The relations developed are limited to polycrystalline surfaces of the metals studied in Houston's experiment. These developments may be made more general by taking note of Houston's plots of T/T_r for a given φ plotted against φ_0. Inasmuch as these plots are nearly linear, an lms fit will produce α and β, where

$$\frac{T}{T_r} = \alpha\varphi_0 + \beta \tag{3}$$

Extending this procedure to obtain two values of φ, say φ_1 and φ_2, would result in

$$\left.\begin{aligned} \varphi_1 &= a(\alpha_1\varphi_0 + \beta_1) + b \\ \varphi_2 &= a(\alpha_2\varphi_0 + \beta_2) + b \end{aligned}\right\} \tag{4}$$

And the solution for a and b as a function of φ_0 would follow immediately.

Good fits can be obtained by using equation (3) for essentially all values of φ within the range ~2.20 to ~3.4 eV (range of interest in all practical power producing thermionic converters). Plots of the data including the Rasor, Warner theory and the lms linear fit for 3.3 and 2.3 eV are shown in figures 1 and 2, respectively. The limitation of the method was determined by extrapolating the lms fit of the Pt group data down to φ_0 = 3.5 eV and comparing these data with Houston's (ref. 17) results for thorium (Th) and hafnium (Hf), which were taken in a separate experiment using the "plasma-anode" technique. Although cathodes having bare work functions of less than 4.3 eV may not be of interest for thermionic converters, it is interesting to note that at φ = 3.3 eV the extrapolation agrees well with data on both Hf and Th, provided that extensions of the linear portions of the φ, T/T_r curves are used. At 2.3 eV, the agreement is good for Hf but not Th. The Rasor, Warner theory shows good agreement with the data at 3.3 eV; however, at 2.3 eV the Rasor, Warner theory does not correlate the data as well as it did at the higher work function.

If we let $\varphi_1 = 3.3$ eV and $\varphi_2 = 2.3$ eV, equations (4) can be solved to yield

$$\left.\begin{aligned} a &= \frac{1}{0.176\ \varphi_0 - 0.158} \\ b &= 3.3 - a(0.702\ \varphi_0 + 0.193) \end{aligned}\right\} \tag{5}$$

From the preceding, it appears that these results would be useful for $2.3 \leq \varphi \leq 3.3$ eV and perhaps for $4.3 \leq \varphi_0 \leq 6.0$ eV for Cs on metal substrates.

As a test of the self-consistency of this procedure, $T/T_r = \alpha_3\varphi_0 + \beta_3$ was fitted at $\varphi_3 = 2.8$ eV. Then $a(\alpha_3\varphi_0 + \beta_3) + b$ was compared with 2.8 eV and found to agree to within ± 0.05 eV for $4.1 \leq \varphi_0 \leq 6.5$ eV. Table II gives the deviations Δ, s using equations (5). They are larger than in table I, but the Δ_{rms}'s are all less than 0.1 eV.

Equations (1) and (2) as written are not useful for converter system design. First, for a given T, T_r, and cathode surface, φ (and hence J) will generally depend on the converter configuration, that is, the interelectrode spacing and wall temperatures. Second, it would be desirable to use the empiricism in cases where the adsorbate is not coming solely from a vapor, for example, where adsorbate is supplied from the substrate for impregnated cathodes.

Close examination reveals that equations (1) and (2) could be ambiguous (φ depends on more than T and T_r) if it is assumed that φ and J are unique functions of T and μ for a given cathode surface, where μ is the cesium arrival rate. If, for example, an electrode is immersed in a gas of temperature T_g, pressure P_g, and particle mass m_g, $\mu = P_g/\sqrt{2\pi m_g k T_g}$. If Knudsen flow conditions apply, $\mu = \mu_0$ = evaporation rate at the cesium reservoir. But if continuous flow conditions apply, $\mu \cong \sqrt{T_r/T}\ \mu_0$. So for a given T and T_r, different μ's and hence different J's are possible.

In order to develop equations (1) and (2) in a generalized form (and consequently to make the usefulness of a's and b's more general) $T_r = T_r(\mu)$ can be written for the case of Houston's experiments. In Houston's experiment, $P_{Cs} = P_g$ and $T_g = T_r$. Heimel's empirical expression (eq. (8) in ref. 23; best fit to experimental data) that makes use of recent vapor pressure data can be approximated for P_{Cs} in the range of Houston's data (area of thermionic interest) in the cesium arrival rate equation to give

$$\mu \cong C \exp^{(-9027/T_r)}$$

so that

$$\frac{1}{T_r} = -\left(\frac{1}{9027}\right) \ln\left(\frac{\mu}{C}\right) \tag{6}$$

where $C = 1.193 \times 10^{30} \overline{T}_r^{-1.02}$, $\overline{T}_r$ is the mean T_r in table XV:2 of reference 24 for the range of arrival rates of interest, the units of μ are particles $(cm^{-2})(sec^{-1})$, and T and T_r are in K. Values of C for use in converter design are given in table III, together with the maximum error in μ caused by the use of $\overline{T}_r$ rather than T_r in C.

Combining equations (1), (2), and (6), we have

$$\varphi = -\left(\frac{aT}{9027}\right) \ln\left(\frac{\mu}{C}\right) + b \qquad (7)$$

and

$$J = 120T^2 \left(\frac{\mu}{C}\right)^{1.2856\,a} \exp(-11\,605\,b/T) \qquad (8)$$

Equations (7) and (8), used in conjunction with equation (5) will be useful whether the cesium arrives from a gas under Knudsen or continuous flow conditions or even if it arrives from the substrate of an impregnated cathode.

Equations (5) and (7) can be used in a practical converter configuration. Rufeh and Lieb (ref. 25) have reported on a variable spacing converter using a (110) oriented vapor-deposited tungsten emitter. They found the bare work function of the emitter to be 4.78 eV. Knudsen's flow condition is assumed. With $C = 2.067 \times 10^{27}$ $(cm^{-2})(sec^{-1})$ (see table III) ($\ln 1/C = 62.90$) and $\varphi = 4.78$ eV, $\varphi(\mu)$ can be determined from equations (5) and (7). The result is shown to be in quite good agreement with the data in figure 3.

The substrate work function can be obtained from cesiated emission data by solving equations (5) and (7) for the bare work function in terms T, μ, and φ:

$$\varphi_0 = \frac{0.158\,\varphi - \frac{T}{9027} \ln\frac{\mu}{C} - 0.714}{0.176\,\varphi + 0.121} \qquad (9)$$

Since cesium vapor pressure tables are normally available for reducing diode data, it is convenient to define an effective reservoir temperature T_r^*; it is the temperature of a pool of cesium which has an evaporation rate $\mu(T_r^* = T_r$ only when $\mu = \mu_0)$. Then equation (9) becomes

$$\varphi_0 = \frac{0.158\ \varphi + \dfrac{T}{T_r^*} - 0.714}{0.176\ \varphi + 0.121} \tag{10}$$

Equations (9) and (10) are expected to be accurate over the same domain as equations (5) and (7), that is, for $2.3 \leq \varphi \leq 3.3$ eV and perhaps for $4.1 \leq \varphi_0 \leq 6.5$ eV.

Equations (9) and (10) can be used as a check on experimental data, like the bare work function reported by Jacobson and Campbell (ref. 26), for example. They list a substrate work function of 4.76 to 4.79 measured in a vacuum diode. It is assumed that μ based on continuum conditions applies since $d/\lambda > 10$. Thus, we can adjust for arrival rate based on μ_0 by $\mu = \sqrt{T_r/T}\ \mu_0$. Setting $T_r = 620$ K at $T = 1916$ K gives an adjusted $\mu = 4.6\times10^{20}$ $(cm^{-2})(sec^{-1})$. For this adjusted μ, $T_r^* = 595$ K. When the value for $\varphi = 2.8$ is used and inserted along with μ and T_r^* into equation (10), it is found that $\varphi_0 = 4.8$. Similar agreement was found with other combinations of d, T, and T_r^*.

This check on diode data is dependent upon the test environments that exist in the cesiated and the vacuum work function studies. Since Cs is a good getter for oxygen, there could be different amounts of oxygen contamination on the electrode surfaces in the cesiated and the vacuum work function tests if the electrode material or the test environment was altered.

The effect of oxygen on cesiated diode performance can be quite strong, as was pointed out as early as 1931 by Villars and Langmuir (ref. 27). This effect appears to be beneficial to thermionic converter performance. As a result, it would be well if the emission properties of the multiple additive thermionic converter could be formulated by a simple relation.

The analysis presented in this report can be used for oxygenated cesiated diodes by altering the bare work function φ_0. Kitrilakis, Lieb, Rufeh, and Van Someren (ref. 28) found that oxygen addition changed the bare-tungsten-surface work function from 4.62 to values between 5.02 and 5.32. Figure (V-1) from reference 29 is used to compare predictions of equations (1) and (5) and of the Rasor, Warner correlation in figure 4. Most of the data for a variety of temperatures (T) are contained within the two lines obtained by analytical estimate from equations (1) and (5). As the figure shows and as Kitrilakis noted, the early Rasor, Warner formulations fail at lower effective work functions.

THE ANODE

The temperature requirements on the anode (<1200 K) are much less stringent than on the cathode. This, of course, gives us much more latitude.

The anode generally has many more surface impurities than the cathode; so Houston's results are not necessarily pertinent to real converter anodes. Still it would improve our understanding if we could predict the minimum work function of "clean" cesiated anodes.

Although the minimum work function φ_{min} is 1.5 ± 0.1 eV for many substrates (refs. 14 and 19) it is desirable to have a means of predicting φ_{min} generally, since we have some freedom in choosing anode materials. Swanson and Strayer (ref. 13) have empirically determined that

$$\Delta\varphi_{min} = 1.09\,(1.78 - \varphi_0) \tag{11}$$

or

$$\varphi_0 + \Delta\varphi_{min} = \varphi_{min} = 1.95 - 0.09\,\varphi_0 \tag{12}$$

Equation (11) was obtained by correlating field-emission data for Cs on molybdenum (Mo), W, W(100), W(110), rhenium (Re), tantalum (Ta), and nickel (Ni). Although thermionic measurements are done at much lower fields and higher temperatures, table IV shows that Houston's results are in good agreement with equation (11). Thus, it appears that we may be able to use equation (12) for thermionic work functions also.

Equation (12) can also be compared with experimental data. Jacobson and Campbell (ref. 27) have plotted their experimental data for φ_{min}, and it is shown in figure 5 along with the prediction of equation (12). Fair agreement is obtained, Jacobson and Campbell data can be correlated by

$$\varphi_{min} = 2.820 - 0.274\,\varphi_0 \tag{13}$$

Several investigators (refs. 29 to 31) have discussed the difficulties involved in determining collector work functions in thermionic converters. Hansen (ref. 30) suggested that the error band for the measured value of collector work function could be 0.2 eV. Rufeh and Lieb (ref. 31) showed similar discrepancies in their measured values of collector work functions when compared with work function values actually achieved in practical regions of thermionic converter operations.

DISCUSSION

The accuracy of our formulation can be compared with that of the Levine (or Steiner), Gyftopoulos and the Rasor, Warner correlations using Houston's data. The Steiner,

Levine, Gyftopoulos (SLG) correlation require knowledge of not only the bare work function but also the monolayer work function and cesium coverage. As Houston did not measure these last two quantities, the SLG correlations unfortunately cannot be compared unambiguously. Houston (ref. 17) compared the calculated (Levine, Gyftopoulos) S-curve properties listed in table 1 of reference 6 with his data. (Table I was made up assuming a monolayer work function of 1.81 eV and a monolayer coverage of 4.8×10^{14} cm^{-2} for Cs on 21 metals.) Houston concluded that "little correlation exists between the experimental results and the theoretical prediction."

The relations derived here do not include the effects of strong electric fields. In taking this into account Langmuir and Taylor (ref. 1) found that such fields affect emission from cesium coated tungsten more strongly than that from pure tungsten - and variably with coverage.

EXAMPLE APPLICATION OF THE FORMULATION

Finally, to illustrate the use of the preceding equations the zero-field electron emission characteristics are calculated for a converter with the following properties:

(1) Bare work function of the emitter, $\varphi_0 = 4.9$ eV

(2) Bare work function of the collector, $\varphi_{0,c} = 5.0$ eV

(3) Emitter temperature, $T_e = 1900$ K

(4) Collector temperature, $T_c = 850$ K

(5) Cesium particle arrival rate at the emitter, $\mu = 1.43\times10^{20}$ $(cm^{-2})(sec^{-1})$

From equations (5), a = 1.42 eV and b = -1.87 eV. Then from equations (7) and (8) we have (using $C = 1.794\times10^{27}$ $(cm^{-2})(sec^{-1})$) $\varphi = 3.02$ eV and $J = 4.35$ $(A)(cm^{-2})$, where φ and J are, of course, the cathode effective work function and electron emission current density, respectively.

If we have a clean collector surface, equation (12) gives its minimum work function as $\varphi_{min} = 1.49$ eV.

CONCLUDING REMARKS

An empirical converter surface physics formulation has been provided which is quite simple and yet agrees better with experiment than other correlations in general use.

Lewis Research Center,
National Aeronautics and Space Administration,
Cleveland, Ohio, June 9, 1971,
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TABLE I. - RESULTS OF LEAST-MEAN-SQUARE FIT FOR a AND b

FROM HOUSTON'S DATA (REF. 19)

Material	Constant, a, eV	Constant, b, eV	rms deviation, Δ_{rms}, eV	Maximum deviation, Δ_{max}, eV	Range in work function φ compared, eV
Pt	1.13	-1.401	0.029	0.044	2.10 to 3.41
Ir	1.33	-1.87	.014	.024	2.04 to 3.45
Os	1.35	-1.83	.014	.033	2.02 to 3.59
Rh	1.42	-2.09	.022	.052	2.31 to 3.60
Ru	1.43	-1.93	.031	.076	2.06 to 3.55
W	1.48	-1.88	.013	.026	2.14 to 3.26

TABLE II. - ACCURACY EVALUATION FOR EQUATIONS (5)

Material	rms deviation, Δ_{rms}, eV	Maximum deviation, Δ_{max}, eV	Range in work function φ compared, eV
Pt	0.099	0.17	2.10 to 3.41
Ir	.035	.058	2.04 to 3.45
Os	.016	.028	2.02 to 3.59
Rh	.089	.17	2.31 to 3.60
Ru	.054	.076	2.06 to 3.55
W	.021	.034	2.14 to 3.26

TABLE III. - ARRIVAL RATE PARAMETER

Cesium particle arrival rate, μ, $(cm^{-2})(sec^{-1})$	Arrival rate parameter, C, $(cm^{-2})(sec^{-1})$	Maximum disagreement with Heimel's μ, percent
3.4×10^{17} - 1.1×10^{19}	2.437×10^{27}	8.9
1.1×10^{19} - 1.4×10^{20}	2.067×10^{27}	7.6
1.4×10^{20} - 8.8×10^{20}	1.794×10^{27}	6.6

TABLE IV. - TEST OF EQUATION (9) UNDER THERMIONIC CONDITIONS

Material	Substrate work function, φ_0^*, eV	Change in substrate work function, $(\varphi_0 - \varphi_{min})^*$, eV	Computed change in substrate work function, $1.09(\varphi_0 - 1.78)$, eV
Pt	5.50	3.9	4.05
Ir	5.31	3.7	3.85
Rh	5.25	>3.7	3.78
Os	5.16	3.7	3.68
Ru	4.89	>3.3	3.39
W	4.69	>3.1	3.17
Hf	3.97	>2.3	2.39

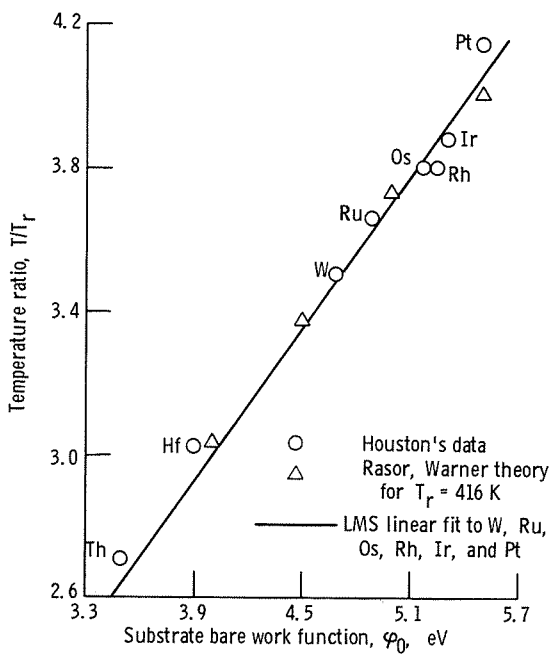

Figure 1. - Temperature ratio as function of substrate bare work function. Effective work function, 3.3 eV.

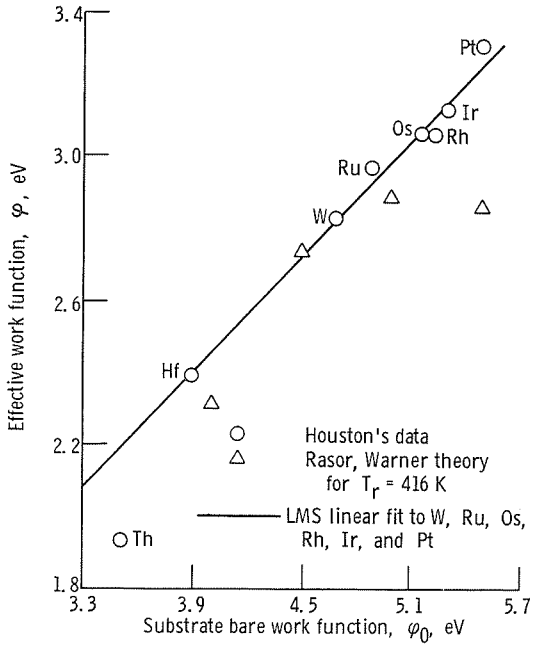

Figure 2. - Temperature ratio as function of substrate bare work function. Effective work function, 2.3 eV.

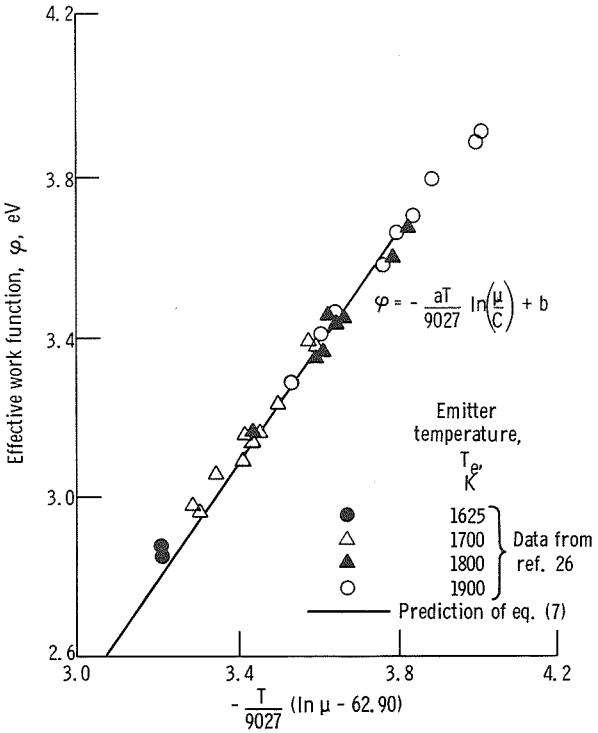

Figure 3. - Comparison of results of reference 25 with predictions of equation (7). Emitter work function, 4.78 eV.

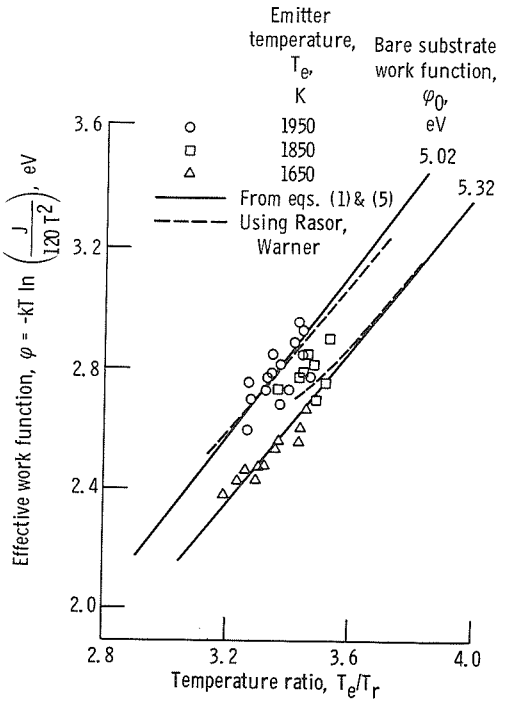

Figure 4. - Experimental and theoretical cesiated-surface work functions with electronegative additive.

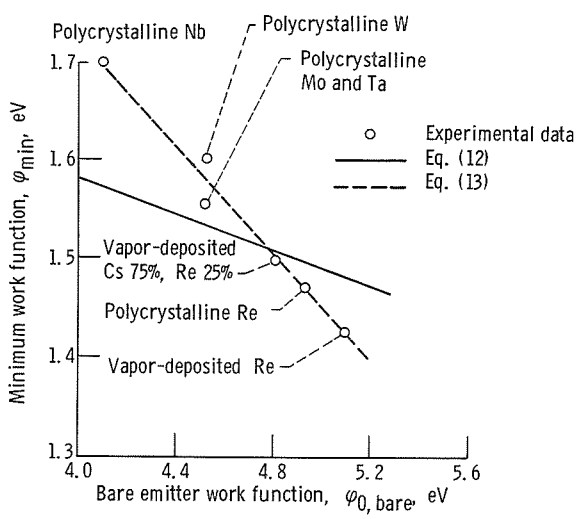

Figure 5. - Test of equation (12).

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